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Volume *versus* surface sampling of neutrons evaporated from hot nuclei

N M Eldebawi
Department of Physics, Faculty of Science, Zagazig University, Egypt

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Abstract : The excitation functions of (heavy ion, xn) reactions are analyzed in terms of an approximate analytical expression for the evaporation cross section consisting of two terms, corresponding to neutron emission from the volume and surface of the compound nucleus. The cooling of the compound nucleus during the evaporation process and the influence of its angular momentum on neutron emission are approximately taken into account. Data on ^{197}Au (^{12}C, xn) and ^{238}U (^{12}C, xn) reactions seem to favor volume sampling for the evaporated neutrons at the lower excitation energies and surface sampling at the higher energies.

Keywords : Heavy ion reactions, compound nucleus, evaporation cross section

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1. Introduction

Energy spectra of neutrons emitted from excited nuclei are often parametrized in terms of a thermal distribution of the form

$$N_\alpha(E, T) = \frac{E^\alpha \exp(-E/T)}{T^{\alpha+1}\Gamma(\alpha+1)}, \tag{1}$$

where E is the neutron energy in the center of mass frame of the emitting nucleus, T is the nuclear temperature, α is a constant exponent and $\Gamma(x)$ is the gamma function. The denominator is introduced to make N_α (E, T) normalized to unity. In the case of charged particle emission, E is multiplied by $(1 - V_C/E)$ here V_C is the Coulomb barrier. Eq. (1) with $\alpha = 1/2$ was first introduced by Weisskopf [1] in his evaporation model. The exponent $\alpha = 1$ has been used for a long time in the phenomenological analysis of evaporation spectra (*e.g.*, ref. [2] and [3]). Spectra of neutrons emitted by heavy-ion reaction products

are often analyzed [4] by assuming that they are coming from two sources : a slow one characterized by the form of eq. (1) with $\alpha = 1/2$ and a faster one with $\alpha = 1$. In the case of charged particle emission, it is doubtful whether $\alpha = 1/2$ or 1 could be distinguished, since barrier effects are strongly energy dependent [5].

Goldhaber [6] pointed out that the pre-exponential factor in the thermal spectrum (1) contains significant information about the nature of the evaporation of neutrons. A pre-exponential factor with $\alpha = 1/2$ is obtained if all the nucleons of the excited nucleus are sampled in some volume of gas in thermal equilibrium. In the other hand, the power $\alpha = 1$ corresponds to a surface sampling which is appropriate for evaporation from the nuclear surface only. In this case, the extra velocity factor arises from the fact that fast particles are more likely to come out in any given time interval.

In this paper, we analyze the excitation functions of (HI, xn) reactions. The probability of emitting x neutrons from both volume and surface sampling of a compound nucleus is obtained by folding x functions of the form of eq. (1). However, different values of T enter in the folded distribution, since the temperature of the compound nucleus decreases after each neutron evaporation. These values depend upon the energies of the previously emitted neutrons. In order to simplify the calculation, we shall replace the values of $N(E_i, T_i)$ by an average $\overline{N}(E_i)$ describing the effective energy spectrum of the cascade neutron. Following Le Couteur [7], we search for this function in the form of a gamma distribution having a mean and a variance chosen to secure an agreement with the results of a simplified evaporation treatment. Angular momentum effects are approximately taken into account by reducing the excitation energy of the decaying nucleus by the amount used up as rotational energy. The cross section of the (HI, xn) reaction is then build up as a superposition of contributions from volume and surface evaporation.

2. Theory

We assume that (HI, xn) reactions proceed through the stage of formation of an equilibrated compound nucleus which de-excites by successive neutron evaporation. Each neutron emission has a spectrum given by eq. (1) which contains contributions from both volume ($\alpha = 1/2$) and surface emission ($\alpha = 1$). During the course of the evaporation process, the temperature of the emitting nucleus monotonously decreases to zero from an initial temperature of :

$$T_{max} = (E^*/a)^{1/2}, \tag{2}$$

where E^* is the excitation energy of the compound nucleus and a is the level density parameter. The probability that x-neutrons are emitted with total kinetic energy

$$E^* - \sum B_i = E^* - x\langle B\rangle$$

where $\langle B \rangle$ denotes the mean binding energy, is given by

$$P_{xn}(E^*, \alpha) = C \int \prod_{i=1}^{x} dE_i \, N_\alpha(E_i, T_i) d\left(\sum_{i=1}^{x} E_i - E^* - x < B > \right), \tag{3}$$

where T_i is the temperature of the residual nucleus after emitting i nucleons which depends on E_1, E_2, ..., E_i. Rather than calculating the emission probability for each neutron at the instantaneous temperature at the moment of emission T_i, we shall perform the calculations after replacing each function $N_\alpha(E_i, T_i)$ by an effective probability of emission. This latter has the form of a gamma distribution with mean value and variance of neutron energy evaluated by the following method, originally proposed by Le Couteur [8].

The mean number of particles emitted, from a nucleus $\bar{n}_\alpha$, with initial excitation energy E^*, is calculated from the integral equation for the cascade [8] :

$$\bar{n}_\alpha = \int^{E^* - \frac{1}{2}<B>} \frac{dE^*}{\langle B \rangle + (\alpha + 1)T}, \tag{4}$$

where $(\alpha + 1)T$ is the mean value of the neutron kinetic energy calculated using eq. (1). The upper limit of the integration is chosen so that $\bar{n}_\alpha = 1/2$ for $E^* = \langle B \rangle$ since the emission of zero or one particle is equally likely. Using eq. (2) together with the dimensionless parameter $S = 2T_{max}/\langle B \rangle$ where T_{max} is the temperature corresponding to the excitation energy $E^* - 1/2\langle B \rangle$, we obtain

$$\bar{n}_\alpha = \frac{E^* - \frac{1}{2}\langle B \rangle}{\langle B \rangle} \; 1 - \frac{2}{3}S + \frac{1}{2}S^2 + \tag{5}$$

The average temperature $\bar{T}_\alpha$ is therefore given by

$$\bar{T}_\alpha = \frac{2}{3} T_{max} - \frac{(\alpha + 1)}{18} \frac{T_{max}^2}{\langle B \rangle} \tag{6}$$

Neglecting the higher order terms in $T_{max}/\langle B \rangle$, we obtain for the average and variance of the emitted neutron kinetic energy the following expressions :

$$\langle E \rangle = (\alpha + 1)\bar{T}_\alpha = \frac{2}{3}(\alpha + 1)T_{max} - \frac{(\alpha + 1)^2}{18} \frac{T_{max}^2}{\langle B \rangle}, \tag{7}$$

$$\sigma^2(E) = T_{max}^2(\alpha + 1)\left(\frac{5}{9} + \frac{1}{18}\alpha\right) + \frac{T_{max}^3}{\langle B \rangle}(\alpha + 1)^2\left(\frac{\alpha - 8}{135}\right). \tag{8}$$

We now search for the mean energy spectrum of the cascade neutrons in the form of a gamma distribution

$$N_\alpha(E) = \frac{E^{t_\alpha - 1}}{\Gamma(t_\alpha)\left(\tau_\alpha^*\right)^{t_\alpha}} \exp\left(-E/\tau_\alpha^*\right). \tag{9}$$

We define the parameters ℓ_α and τ_α^* of the distribution (9) by requiring the energy mean and variance to be equal to the ones given by eqs. (7) and (8) respectively. We thus obtain

$$\ell_\alpha = \frac{8(\alpha+1)}{\alpha+10} \quad \text{and} \quad \tau_\alpha^* = \frac{T_{\max}}{12}(\alpha+10). \tag{10}$$

The results obtained by Le Couteur [7,8] correspond to the choice $\alpha = 1$ in eq. (1) *i.e.*, to surface sampling. In this case,

$$\ell = \frac{16}{11} \quad \text{and} \quad \tau^* = \frac{11}{12} T_{\max}. \tag{11}$$

For volume sampling *i.e.*, $\alpha = 1/2$ in eq. (1), the equivalent quantities are given by

$$\ell = \frac{8}{7} \quad \text{and} \quad \tau^* = \frac{7}{8} T_{\max} \tag{12}$$

Now we replace the functions $N(E_i, T_i)$ in eq. (3) by $\bar{N}_\alpha(E_i)$, as given by eq. (9), and change the integration variable E_i to r_i^2, to obtain

$$P_{xn}(E^*, \alpha) = C\left[\frac{1}{2}(\tau_\alpha^*)^{\ell_\alpha}\,\Gamma(\ell_\alpha)\right]^{-x} \exp\left(-\frac{E^* - x\langle B\rangle}{\tau_\alpha^*}\right)$$

$$\int_0^\infty r_1^{2\ell_\alpha - 1}\, dr_1 \cdots \int_0^\infty r_x^{2\ell_\alpha - 1}\, dr_x\, \delta\left(r_1^2 + \cdots r_x^2 - R^2\right), \tag{13}$$

where $R^2 = E^* - x\langle B\rangle$. The integral in eq. (13) can be expressed in terms of surface areas of hyper spheres in the 2ℓ- and 2α-dimensional spaces [9]. One then obtains

$$P_{xn}(E^*, \alpha) = C\left(\frac{E^* - x\langle B\rangle}{\tau_\alpha^*}\right)^{\ell_\alpha x - 1} \exp\left(-\frac{E^* - x\langle B\rangle}{\tau_\alpha^*}\right) \Big/ \left(\tau_\alpha^* \Gamma(\ell_\alpha x)\right) \tag{14}$$

We have introduced the constant C in order to change the normalization so that

$$\sum_{x=1}^{x_{\max}} P_{xn} = 1, \tag{15}$$

where $x_{\max} = E^*/\langle B\rangle$. Replacing the summation in eq. (15) by an integration and using the method of steepest descent yields [10]

$$C = \ell_\alpha \tau_\alpha^* + \langle B\rangle. \tag{16}$$

We shall assume that the number of neutrons evaporated from each of the volume and surface region of the compound nucleus is proportional to the excitation energy deposited in the corresponding domains. The decomposition of the excitation energy into volume and surface contributions is made following the liquid drop model approach of Stocker and Burzlaff [11]. These authors introduce "a density dependent expression for the interaction energy which implicitly depends upon temperature by means of

a temperature dependent nuclear equilibrium density". Following their treatment, we write

$$a = a_v A + a_s A^{2/3}, \tag{17}$$

where A is the mass number of the compound nucleus and a_v and a_s are the level density parameters for the volume and surface parts of the nucleus.

The cross section of a (HI, xn) reaction, when both volume and surface sampling contribute to the process of neutron evaporation, can be written as

$$\sigma_{xn} = \sigma_{cf} P(E^*, \alpha)\, \Theta\left(E^* - x\langle B\rangle\right), \tag{18}$$

where σ_{cf} is the cross section for the complete fusion of the target and projectile. The theta function $\Theta\left(E^* - x\langle B\rangle\right)$ expresses the fact that the binding energies of x-neutrons in the compound nucleus cannot exceed its excitation energy E^*. The factor $\left(\overline{\Gamma_n/\Gamma}\right)^x$ accounts for the competition of neutron evaporation with fission and charged particle emission. It was first introduced by Sikkeland *et al* [12]. In principle, a factor $\left(\Gamma_n/\Gamma\right)$ is introduced for each step of the evaporation cascade and depends upon excitation energy of the intermediate nucleus, its mass and atomic weight because of the pairing and shell effects. Replacing the products of the branching ratio by a power of their mean value is an approximation which might be justified by the relatively high mean temperature (~1.5 – 2.0 MeV) involved during the evaporation cascade.

The fusion cross section can be calculated following Glas and Mosel [13],

$$\begin{aligned} \sigma_{cf} &= \pi R_a^2 \left(1 - V_C/E\right), && E < E_{GM}; \\ &= \pi R_f^2 \left(1 - B_f/E\right), && E < E_{GM}; \end{aligned} \tag{19}$$

where $E_{GM} = \left[V_C\left(R_a/R_f\right)^2 - B_f\right] \Big/ \left[\left(R_a/R_f\right)^2 - 1\right]$ while B_f and R_f are the fusion barrier and radius, respectively, which can be taken from Birkelund and Huizenga [14]. V_C is the Coulomb potential at the strong absorption radius $R_a = 1.5\left(A_1^{1/3} + A_2^{1/3}\right)$ fm.

3. Angular momentum effect

In principle, the angular momentum acquired by the compound nucleus at the onset of the reaction, should affect all the quantities involved in eq. (18). One may then assume that the cross section of (HI, xn) reactions is expressed as

$$\sigma_{xn} = \sum \sigma_{xn}(J), \tag{20}$$

where the terms $\sigma_{xn}(J)$ are the cross sections for emitting x-neutrons from a compound nucleus with spin J. Due to the large values of angular momentum involved in the

reactions, we neglect the spins of the target and projectile so that the angular momentum of the compound nucleus J is taken to be the orbital momentum of the initial channel. The independence of the formation and decay of the compound nucleus enables us to write

$$\sigma_{xn}(J) = \sigma_{cf}(J)\left(\overline{\Gamma_n/\Gamma}\right)^x P_{xn}(E^*, \alpha, J), \tag{21}$$

where $\sigma_{cf}(J)$ is the cross section of complete fusion of target and projectile at relative angular momentum J,

$$\sigma_{cf}(J) = \pi\lambda^2(2J+1)T_J. \tag{22}$$

λ is the incident reduced wavelength and T_J the transmission coefficient which is usually evaluated by varying the parameters of the parabolic barrier approximation used in a statistical model computer code to fit the σ_{cf} data. We use for T_J, the sharp cut-off approximation which proved to be quite successful [15] :

$$T_J = \begin{matrix} 1 & \text{for} & J < J_{lim} \\ 0 & \text{for} & J > J_{lim} \end{matrix}.$$

Angular momentum also affects the probability of x neutron emission $P_{xn}(E^*, \alpha, J)$ and the factor governing the competition between fission and neutron emission $\overline{\Gamma_n/\Gamma}$. In the present formulation, we have replaced the branching ratio Γ_n/Γ by its mean value $\overline{\Gamma_n/\Gamma}$, where the averaging is over all the x neutron emission channels. In different channels, the cross sections have peaks at different energies and thus involve different ranges of orbital angular moment. Thus, we expect the averaging over x to smooth out the angular momentum dependence of $\overline{\Gamma_n/\Gamma}$ and consider this quantity as being calculated at a certain average value of angular momentum depending on the value of x. We shall indeed see that the best-fit values of $\overline{\Gamma_n/\Gamma}$ are more dependent on the number of emitted neutrons in the case of the heavier target (see Table 1). Since the moment of inertia of the highly distrorted neucleus at saddle is larger than that of a spherical nucleus at equilibrium,

Table 1. Value of $\overline{\Gamma_n/\Gamma}$ used in the calculation.

	^{197}Au (^{12}C, xn)		^{38}U (^{12}C, xn)	
x	$\overline{\Gamma_n/\Gamma}$ Surface	$\overline{\Gamma_n/\Gamma}$ Volume	$\overline{\Gamma_n/\Gamma}$ Surface	$\overline{\Gamma_n/\Gamma}$ Volume
4	0.65	0.71	2	2.8
5	0.62	0.67	1.2	1.3
6	0.65	0.67	0.77	0.77
7	0.64	0.62	0.61	0.61
8	0.82	0.76	0.48	0.48

therefore a smaller amount of energy is tied up in rotational at saddle than at equilibrium shape, which decreases the height of the fission barrier [16]. Hence, fission competes more

favorably with neutron emission as the angular momentum of the system is increased [16]. Finally, angular momentum affects the probability of emission of x neutrons $P_{xn}(E^*, \alpha, J)$ at an excitation energy E^* since the latter is effectively decreased by an amount given by the rotational energy, we thus write :

$$\sigma_{xn} = \left(\overline{\Gamma_n/\Gamma}\right)^x \sum_{J=0}^{J_{\lim}} \sigma_{cf}(J)\, P(\tilde{E}^*, \alpha)\, \Theta\left(\tilde{E}^* - x\langle B\rangle\right), \tag{23}$$

where we replace $P_{xn}(E^*, \alpha, J)$ by $P_{xn}(\tilde{E}^*, \alpha)$ given by eq. (14) with the excitation energy available E^* being replaced by

$$\tilde{E}^* = E^* - E_R = E^* - h^2 J(J+1)/2I, \tag{24}$$

where E_R is the rotational energy of the compound system characterized by a moment of inertia

$$I = (2/5)\, m r_0^2 A^{5/3}. \tag{25}$$

The upper limit of the summation in eq. (23), $J_{\lim}$ is the critical angular momentum above which no compound nucleus is formed [17]. It is taken to be the lower of the two values :

$$\begin{aligned} J_{\lim} &= kR_f\sqrt{1 - B_f/E_i}, \\ J_{\lim} &= kR_a\sqrt{1 - V_c/E_i}, \end{aligned} \tag{26}$$

where R_f, B_f, R_a and V_c are the quantities already defined in eq. (19). Since the quantity $\overline{\Gamma_n/\Gamma}$ is taken as a fitting parameter, therefore its dependence upon angular momentum is inclusive in the parametrization choice.

4. Comparison with experiment and discussion

We used eq. (23) to calculate the cross sections for the reactions ^{197}Au (^{12}C, xn) and ^{238}U (^{12}C, xn) by including both volume ($\alpha = 1/2$) and surface evaporation ($\alpha = 1$) in eq. (1). These choices for α for a single neutron emission corresponding to $l = 16/11$ and $\tau^* = (11/12)T_{max}$ for surface sampling, and to $l = 8/7$, $\tau^* = (7/8)\, T_{max}$ for volume sampling, as deduced in eqs. (11, 12). The value of $\overline{\Gamma_n/\Gamma}$ used in the calculation are given in Table 1. The level density parameter a is evaluated according to eq. (17). Figure 1 shows a comparison between the results of calculations with the experimental data of references [18,19]. The figure shows that the calculations with volume evaporation are in good agreement with the data in a narrow range of excitation energies just above the corresponding reaction threshold. An exception is the case of 4- and 5-neutron emission in the interaction with the ^{238}U target because the thresholds are at energies below the Coulomb barrier where the sharp-cutoff model yields zero reaction cross section. Relaxing the sharp cutoff will not improve the agreement by much, since as well-known, barrier penetration models cannot account for sub-barrier fusion [20]. We also see this from the fact that the obtained values of $\overline{\Gamma_n/\Gamma}$ are more than 1. At higher excitation energies, on the

other hand, surface sampling leads to a better description of the data especially for reactions involving more neutron evaporation.

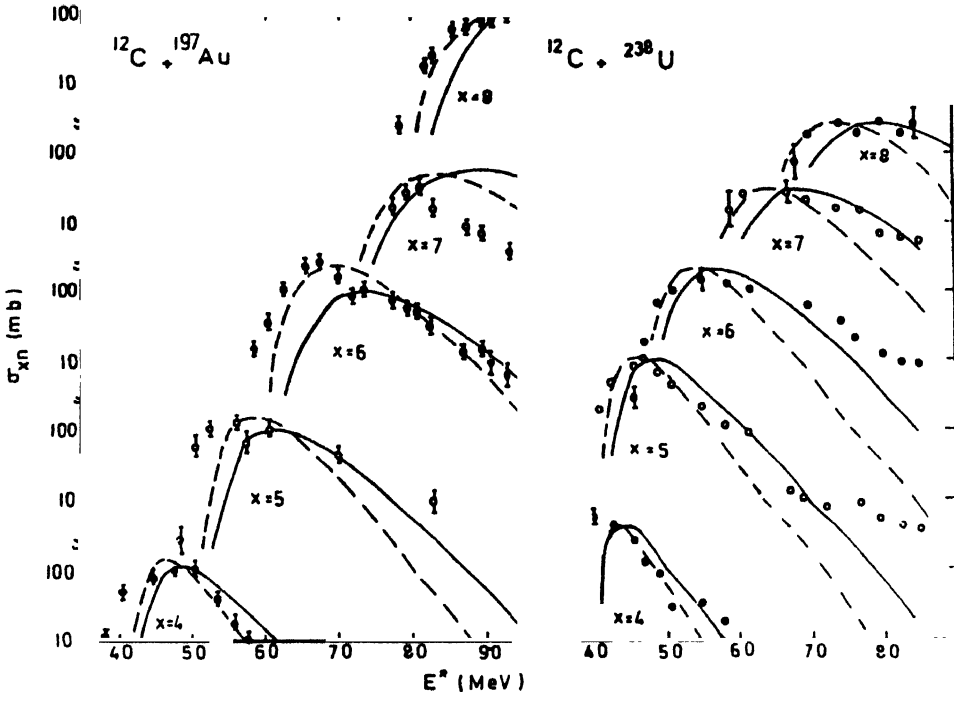

Figure 1. Excitation functions for ^{197}Au (^{12}C, xn) and ^{238}U (^{12}C, xn) reactions. The dots are the experimental data of Refs. [18] and [19] respectively. The curves are calculated using eq. (23) with $P_{xn}(\tilde{E}, \alpha, J)$ from (14) and (24). The solid curves represent surface sampling ($l = 16/11$, $\tau^* = (11/12)T_{max}$), while the dashed curves represent volume sampling ($l = 8/7$, $\tau^* = (7/8)T_{max}$)

In order to make definite conclusions about whether the neutron emission happens from the nuclear volume or surface, one has to perform numerical calculations using codes based on more elaborate evaporation theories. However, the large difference between the outcome of the surface and volume samplings shown in the present paper encourages one to believe that the conclusion drawn above on the basis of the approximate formulation is valid. Moreover, the analytical results obtained here show that the decisive factor in deciding about the volume or surface character of the neutron emission is the position of the peak of the excitation functions. Eq. (18) shows that if one neglects the dependence of σ_{xn} on E^* near the peak, which is true except for small values of x, the maximum of σ_{xn} occurs at $E^*_{peak} = x\langle B\rangle + (xl-1)\tau^*$ yielding :

$$E^*_{peak} = (x+1)\langle B\rangle + \left(\frac{4}{3}x - \frac{11}{12}\right)T_{max}, \text{ for surface sampling}$$

$$= (x+1)\langle B\rangle + \left(x - \frac{7}{8}\right)T_{max}, \text{ for volume sampling.}$$

This equation can thus be used to get a rapid estimate from the peak position about whether volume or surface sampling prevails.

We can support the conclusion that neutron evaporation at relatively high excitation energies is mainly a surface effect by the fact that phase transitions in liquid drops occur predominantly at the surface. Indeed, it has been demonstrated both experimentally [21] and theoretically [22] that the melting process starts from the surface layer and propagates into the interior and that the surface melts at temperatures significantly lower than the bulk melting point.

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